Replies to the comment of anonymous referee 2

The paper in question is a new effort to tighten up the atmospheric molecular hydrogen budget with a high resolution global chemistry and transport model, TM5. The paper does suffer from the fact that it reads like a mixture of carefully detailed photochemical model development and arbitrary changes in various source and sink strengths/fractionations in order to adjust model results to match observations. Nevertheless, it is a welcome piece of work that is worthy of publication.

[We regret that our line argumentation behind applying the changes made in the source and sink strengths is not clear. It was known in advance that the fossil fuel related emissions, originally derived from CO emissions and known H_2/CO ratios, were underestimated by the GEMS dataset. Using previously reported estimates for fossil fuel related H_2 emissions to rescale the dataset is therefore in our view justifiable before starting a comprehensive study. After first investigatory calculations it also appeared that deposition was overestimated by the model (more pronounced at the NH), so again the sensible thing to do was to look at the uncertainties in the deposition parameterisation to look for justifiable room to alter the overall magnitude of the parameterisation. Apart from the emissions and deposition, nothing else was altered. The chemistry model was used in its original form (Pieterse et al., 2009). We took over the isotope signatures from existing literature, again a justifiable choice we think. The final objective of our efforts to build this new model framework in TM5 is of course to perform an inverse calculation to quantify the impact of the different processes on the global and regional scale. The manual adjustment of the driving parameters within the reported ranges of uncertainty to obtain first reasonable results is basically the first step towards a full inverse model study.]

Page 5813, line 9. Should also reference Jacobson, either Science or GRL.

[Indeed. we will add the reference for the 2005 Science paper.]

Page 5813, line 21. References needed. The 1.4 year lifetime comes from Rhee et al., a piece of work that this paper apparently discredits and the large variation in current estimates of H_2 lifetime should not be emphasized simply in order to validate the current work.

[It was not our intention to use this range to justify the correctness of our results or to narrow down the uncertainty of the current estimates. The main goal was to implement the photochemistry and see how the resulting framework would perform. This is also the reason why we chose not to close the budget by tweaking one/more parameters to values that cannot be validated at present. The secondary goal was to identify the largest uncertainties/unknown parameters in the isotope budget so that future work can concentrate on these likely most important issues. We agree that the lifetime of the Rhee paper is likely too low, and will discuss that in detail in a companion paper (Batenburg et al., 2011), see also reply to last point below]

Page 5821, lines 12-16. If the discrepancy between model and measurements were due to measurements being made in clean air, then the difference between the two should be greatest for highly populated northern latitudes and should trend toward zero at high latitudes but this is not the case. If anything the data appear to show the opposite.

[The referee is correct, the term clean background air is inappropriate, we will change this sentence. We do not expect that the differences between the model and measurements should be smaller for the higher NH latitudes. For the NH, deposition and surface sources dominate all other photochemistry, leading to overall lower mixing ratios

(deposition overrules emissions in the large scale signals) and isotopic composition over land than over the oceans (location of many observational sites). The sensitivity study also shows little sensitivity to relatively large changes in the surface source signatures. Thus, on average the model will underestimate the observational sites, which is shown in the figure. In the tropics, the spatial and temporal variability is very large, due to biomass burning and fossil fuel related emissions but also due to regions of very little activity (i.e. oceans). It is for this reason that we actually expect both, the closest resemblance between the model for regions above the ocean, but also large excursions when the air mass sampled is of continental origin.
But indeed, the most severe issue is observed at the Southernmost latitudes, where we cannot close the gap between the model and measurements by changing emissions. We have to make significant changes in the photochemistry or deposition to match the observations whereas only relatively small changes are necessary in the isotopic composition at the tropopause. This is another reason why we investigate this process in detail. We did not mean to dismiss the discrepancies between model and measurements by mentioning the argument of representativeness of the location of the measurement sites so will reformulate this sentence. However, we do dedicate a large part of the manuscript in trying to find the most likely parameterisation to alter to get the model closer to the observations, but for reasons of completeness we believe it should be mentioned.]

Page 5824, lines 19 - 20. This could also be explained by an increase in production from tropospheric methane which results in isotopically enriched H_2, an increase that would be expected during peak production of OH in the SH summer.

[Indeed. We looked at the fields of for instance HCHO, HO_2 but the locations of the peak values in these fields did not match the location of the peak values in the isotopic composition (the former more to the north than the latter). Also the seasonal difference plots for case 1a/1b (because we had to make choices to cut down the number of plots only the annual mean for 1b is shown in Fig. 9) also showed a spatial disconnection from the observed pattern. This leaves the STE as the most likely candidate to explain this pattern. Cases 2b/2c also indicate that it is indeed the STE that is the most likely candidate.]

Page 5830, lines 20-30. The large proposed mid-latitude strat/trop exchange is somewhat troubling. Some mass balance considerations comparing the mid-lat to high-lat exchange would be welcome. The authors even admit on the next page that TM5 does a poor job with downward transport.

[The first referee also noticed this. Please refer to our response to his remarks for a detailed rebuttal. In fact, the stratospheric contribution in consists of two parts in this model domain. With respect to the downward transport: It is the underlying meteorological data (ECMWF) that overestimates downward transport and herewith any CTM using this data will suffer from similar problems.]

Page 5533, lines 5-8. The authors state that an alpha of 0.9 for soil uptake is out of the range of reported values. At least one of the authors however has seen evidence at least once that there is some evidence that alpha for soil uptake actually is in some cases as great as 0.9 (Rahn, AGU fall meeting 2005). Although not in the peer reviewed literature, the authors reference at least one non-peer reviewed internal report so I see no reason not to either reference the detailed AGU abstract or at least contact the author for permission to use personal communication. It is a rather important parameter in their model and to dismiss the possibility that soil uptake may be solely responsible for the modeled/measured delta based on the meager data available in the literature does a disservice to the reader.

[An appropriate reference will be included in the final version of the paper.]

Finally I cannot help commenting that the current work repudiates in several aspects the earlier work of some members of this team (Rhee et al, 2006) and it would be fitting for the authors to take the opportunity to address the discrepancies between the two pieces of work.

[We will clearly address the discrepancies to the Rhee et al. paper in the revised version, because we agree that we cannot close the H_2 and isotope budget with several of the parameters postulated there (in particular the very high fraction of the soil sink, and combined with this the low atmospheric lifetime, see comment on lifetime above).]